ORIGINAL ARTICLE

Effect of High-Molecular-Weight Sodium Alginate on the Viscosity and Characteristics of Alginate Impression Materials

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Clinical significance

Alginate impression materials are unsuitable for obtaining a preliminary impression for complete dentures because of their low viscosity. Our study suggested that the addition of high-molecular-weight sodium alginate to an alginate impression material can be effective for increasing the viscosity without deterioration of the properties of the material.

Abstract

Purpose: In this study, we investigated the effects of addition of high-molecular-weight sodium alginate to alginate impression materials with the aim of developing a new high-viscosity impression material.

Methods: Four experimental alginate impression materials were prepared based on a conventional material (control material) by varying the percentage of addition of high-molecular-weight sodium alginate in the range of 50% to 100%. The viscosity after mixing, setting time, recovery from deformation, strain in compression, compressive strength, and compatibility with dental stones was determined in accordance with the ISO 1563 and ANSI/ADA Specification no. 18. Results were analyzed using ANOVA and linear regression.

Results: All the experimental materials exhibited significantly higher viscosity than the control material ($P<0.05$). A significant and positive relationship was observed between the addition rate and the viscosity ($P<0.05$). In the specification tests, all the experimental materials were found to be within specification limits, and not significantly different from the corresponding values of the control material.

Conclusion: The addition of high-molecular-weight sodium alginate to an alginate impression material can be effective for increasing the viscosity of the material without deterioration of the properties of the material.

Introduction

For obtaining the preliminary impression for complete dentures, recording the anatomical forms of the alveolar ridge for determining the outline of the denture base area is required.[1] The impression material used must push away the surrounding tissues to a certain extent in order to capture the anatomical form of the alveolar ridge. Therefore, to achieve their purpose, impression materials used for preliminary impressions need to have a high viscosity.

Alginate impression materials are used most commonly for preliminary impressions.[2] Nevertheless, these materials do not have sufficient viscosity to push away the surrounding tissues as mentioned above.[3-5] Therefore, the viscosity of alginate impression materials is usually increased by decreasing the water per powder (W/P) ratio.[6] However, decreasing the W/P ratio results in numerous problems, such as deterioration of the flexibility and elasticity of the material[7,8], difficulty in manipulation, and shortening of the setting times.[9] Therefore, it is necessary to examine the base constituent of alginate impression materials for the purpose of increasing their viscosity.

Alginates (sodium alginate and potassium alginate) are metallic salts of alginic acid, which is a linear polymer of anhydro-β-D-mannuronic acid, and the chief active ingredient of alginate impression materials. When alginates are mixed with water, they form a sol. The viscosity of the alginate sol can be increased by the addition of

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fillers, such as diatomaceous earth.[10] However, addition of excessive quantities of the filler material can cause loss of elasticity, which is the most important property of elastic impression materials.[8] High-molecular-weight alginate also shows a high viscosity of the sol.[11] However, little information is available on the relationship between the viscosity of alginate sol and the molecular weight of the alginate material.

In this work, high-molecular-weight alginate was added to alginate impression materials in order to increase their viscosity. The viscosity of the experimental impression materials and their applicability as impression materials in the field of dentistry were investigated.

Materials and methods

Materials

Alginate impression materials with four experimental compositions were prepared using the basic constituent of ALFLEX (ALF; Nissin, Kyoto, Japan). The contents of alginate in the four materials are given in Table 1. The viscosity of a 1wt/vol% aqueous solution of sodium alginate, used in ALF (molecular weight approximately 300,000-500,000) at 20°C, was 0.50-0.60 Pa·s, while that of high-molecular-weight sodium alginate (HWSA; Nissin, Kyoto, Japan, molecular weight approximately 550,000-850,000) at the same temperature was 1.00-1.10 Pa·s. The compositions of all of the experimental materials were exactly the same, except for the content of alginate.

Methods

The mixing of the impression materials with water was performed with a vacuum-mixing machine (Super Rakuneru, GC, Tokyo, Japan) for 10s. The mixing ratio was 16g of powder with 40ml of water, as recommended by the manufacturer of ALF. The water temperature used for the mixing was 23±1°C. The entire experimental procedure was carried out in an environment with a temperature of 23±2°C and relative humidity of 50 ± 10%.

Viscosity of the alginate sol was measured by a torsional oscillation-type viscometer (VM −100A, CBC Co., Tokyo, Japan). The measurement was performed at 60, 75, and 90 s after the commencement of mixing. The measurement was carried out five times for each material.

Table 1 Content of alginate (mass%).

Material	HWSA	Sodium alginate	Potassium alginate
ALF	0	50	50
EM1	50	0	50
EM2	75	0	25
EM3	90	0	10
EM4	100	0	0

Setting time was measured in accordance with the ANSI/ADA specification no.18 for alginate impression materials.[12] A metal ring, 30 mm in diameter and 16 mm in thickness, was placed on a flat glass plate and filled with the mixed alginate impression material. Excess impression material was leveled off with the top of the ring. The flat end of a polished rod made of poly (methyl methacrylate), 6 mm in diameter and 100 mm long, was then placed in contact with the surface of the impression material, immediately withdrawn, and then cleared of any stuck material. The contact/withdrawal steps were repeated until the rod separated out from the material. The test was carried out in triplicate for each material.

Recovery from deformation, strain in compression, compressive strength and compatibility with dental stones were determined in accordance with the ISO specification 1563 for dental alginate impression materials.[13]

Recovery from deformation was measured as follows. Three specimens of each material were made into cylinders 20 mm high having a diameter of 12.5 mm. A lightweight plate was placed on top of the specimen. The specimen was placed in the deformation apparatus with a dial indicator and the foot of the indicator came into contact with this plate 45 s after the setting times stated by the manufacturer of ALF. The force exerted by the spindle of the indicator was 0.6±0.1 N. This was applied for 10 s before obtaining reading A (mm). Subsequently, the specimen was deformed to a height of 16±0.1 mm (20% strain) and maintained for 5±0.5 s before the deformation force was released. The specimen was allowed to stand for 30 s and the foot of the indicator was then lowered onto the plate for 10 s. The value at this point represented reading B (mm). The recovery from deformation was expressed as a percentage using the following formula:

Recovery from deformation (%)
$$=100\times\{1-(A-B)/20\},$$

where 20 is the length of the specimen, in millimeters.

Strain in compression was measured as follows. The specimens were prepared in a manner similar to that for the test of recovery from deformation. The specimen was placed on the plate of the compression device and subjected to a load of 125±10 g (approximately 0.01 N/mm^2) at 60 s after the setting time stated by the manufacturer of ALF. Thirty seconds later, the value shown by the dial indicator was read as reading C (mm). Sixty seconds after the application of the load of 125±10 g, the load was increased over 10 s to 1250±10 g. Thirty seconds after application of the load of 1250±10 g, a reading of the dial indicator was taken; this value represented reading D (mm). The strain in compression was expressed as a percentage using the following formula:

Strain in compression (%)=100×{(C−D)/20},

where 20 is the length of the specimen, in millimeters.

Compressive strength was measured as follows. The specimens were prepared in a manner similar to that for the test of recovery from deformation. Sixty seconds after the setting time stated by the manufacturer of ALF, the specimen was placed in the universal testing machine (Instron 5544, Instron Co., Norwood, MA). The specimen was loaded continuously and as uniformly as possible to obtain an average rate of loading of 100±20 N/min until fracture. The compressive strength expressed in MPa, was calculated using the following formula:

Compressive strength (MPa)=4F/πd^2,

where F is the force at fracture, in Newtons, and d the diameter of the specimen, in millimeters.

Compatibility with dental stones was determined by measuring the detail reproduction and surface roughness. The dental stone casts made from the impression materials were prepared using a ruled test block as specified in the ISO 1563. This test block has a series of three parallel lines intersected by two fiducial lines at right angles and 25 mm apart. These lines are 20, 50 and 75 μm in width.

Detail reproduction was determined as follows. After mixing the alginate impression material, it was poured into the ring mould, slightly overfilling it. The test block was then centered above the mould and pressed down on the mass of the impression material. Immediately thereafter, it was placed in the water bath at 35±1°C and loaded with 10N. Three minutes after the setting time, the ring mould was separated from the test block. A powder of dental stone (NEW DIASTONE, Ryoka Dental, Mie, Japan) was mixed into water at the water/powder ratio recommended by the manufacturer, and mechanically mixed with a vacuum-mixing machine (Super Rakuneru) for 10 s. The mixture was then poured over the impression material under gentle vibration. The poured impression was allowed to stand for 30 min in excess of the setting time. The dental stone cast was removed from the ring mould containing the impression material, the cast was then observed under low-angle illumination at a magnification of 4× to 12×, and the finest line reproduced over the full length of 25 mm between the intersection lines was recorded. The test was conducted in triplicate for each impression material. The line reproduced by at least two casts from three tests was taken as the test result.

Surface roughness of the dental stone casts from the alginate impression materials was determined using a Profilometer (Surfcom, Tokyo Seimitsu, Tokyo, Japan). The stone casts made for the above test of detail reproduction were used. The mean roughness values (Ra) were measured as the centerline average, in microns. Three measurements were carried out for each stone cast, i.e. 9 measurements for each material.

Statistical analysis

Differences in the viscosity, setting time, recovery from deformation, strain in compression, compressive strength and surface roughness for the five groups were analyzed by one-way ANOVA and Tukey's multiple range tests. In addition, in order to examine the possible correlation and establish regression models between the viscosity and the percentage of addition of HWSA, simple linear regression analysis was applied. All data were analyzed using SPSS version 10.0J (SPSS Japan Inc., Tokyo, Japan) and the probability level was set at $P<0.05$.

Results

The results of the viscosity test are presented in Table 2. All experimental materials containing HWSA had a significantly higher viscosity than ALF, except for EM1 at 90s ($P<0.05$). EM3 and EM4 had significantly higher viscosity than EM1

Table 2 Mean (SD) viscosity (Pa・s) of the alginete impression materials tested after mixing.

Material	ALF	EM1	EM2	EM3	EM4
60 s	3.238 (0.196)[a]	3.610 (0.198)[b]	3.840 (0.110)[b,c]	4.090 (0.102)[c,d]	4.152 (0.141)[d]
75 s	3.482 (0.164)[a]	3.784 (0.197)[b]	4.014 (0.131)[b,c]	4.198 (0.112)[c]	4.262 (0.087)[c]
90 s	3.522 (0.149)[a]	3.822 (0.204)[a,b]	4.050 (0.198)[b,c]	4.220 (0.123)[c]	4.286 (0.097)[c]

N=5 for each test group.
Within a row, groups superscripted with similar lower case letters showed no statistically significant differences ($P > 0.05$).

Table 3 Model summary of the regression analysis.

Time	Regression Equation	r	r^2	P value
60 s	y=0.0093x+3.2013	0.915	0.837	$P < 0.0001$
75 s	y=0.0079x+3.4479	0.904	0.817	$P < 0.0001$
90 s	y=0.0077x+3.4940	0.882	0.778	$P < 0.0001$

at any measurement point ($P < 0.05$).

The relationship between the percentage of addition of HWSA and the viscosity at each measurement time are shown in Figure 1, Figure 2, and Figure 3. The regression model, the correlation coefficient (r), the coefficient of determination (r^2), and the probability level (P-value) are presented in Table 3. Significant correlations were observed between the percentage of addition of HWSA and the viscosity ($P < 0.001$), and the correlation r^2 was high at any measurement point. The correlation r^2 was especially high at 60s after the commencement of mixing ($r^2 = 0.837$), provided that the correlation was strong.

Setting time, recovery from deformation, strain in compression, compression strength, detail reproduction, and the surface roughness of each material are summarized in Table 4. The setting time of all the materials tested in this study was within the ANSI/ADA specification (60 s−300 s). The values of the other parameters mentioned above, except for strain in compression, were greater than the lower limit of the ISO specification for all the tested materials (recovery from deformation, 95%; compressive strength, 0.35 MPa; detail reproduction, 50 μm) and the values of strain in compression were within the range of the ISO specification (5%−20%). EM2 exhibited a significantly higher value of recovery from deformation than ALF ($P < 0.05$). EM1 and EM2 exhibited significantly higher values of compressive strength than ALF ($P < 0.05$). The results of ANOVA indicate no significant differences among the materials in terms of the strain in compression and surface roughness ($P > 0.05$). All materials reproduced the 50 μm-lines, and failed to record the 20 μm-lines.
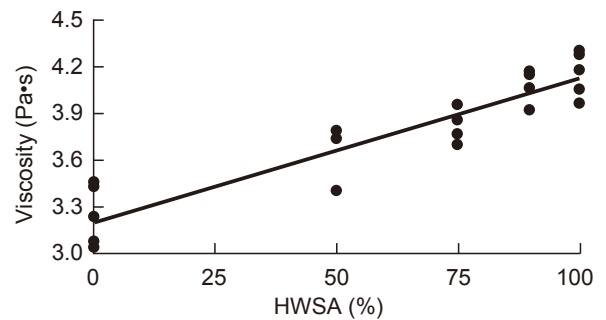

Fig. 1 Correlation between additive rate and viscosity at 60s after mixing started.
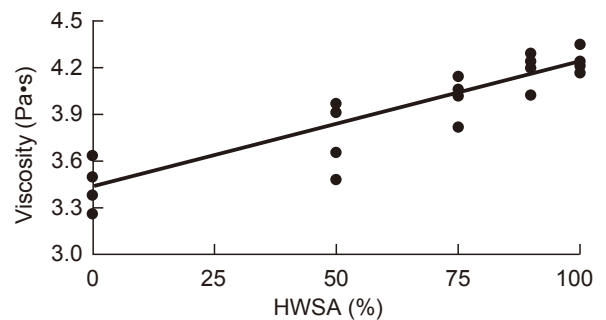

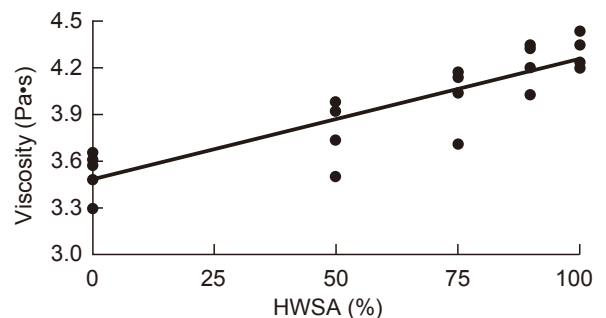**Fig. 2** Correlation between additive rate and viscosity at 75s after mixing started.

Fig. 3 Correlation between additive rate and viscosity at 90s after mixing started.

Table 4 Mean (SD) setting time and physical properties of each material.

	Material				
	ALF	EM1	EM2	EM3	EM4
Setting time (s)	115.0 (0.0)[a,b]	116.7 (2.9)[a]	110.0 (0.0)[b]	111.7 (2.9)[b]	111.7 (2.9)[b]
Recovery from deformation (%)	96.57 (0.33)[a]	96.72 (0.15)[a]	97.60 (0.84)[b]	96.98 (0.57)[a,b]	97.18 (0.01)[a,b]
Strain in compression (%)	16.45 (1.05)[a]	16.75 (0.78)[a]	16.20 (0.63)[a]	16.80 (0.51)[a]	18.00 (1.14)[a]
Compressive strength (MPa)	0.577 (0.017)[a]	0.651 (0.012)[b,c]	0.711 (0.026)[c]	0.590 (0.013)[a]	0.614 (0.022)[a,b]
Surface roughness Ra (μm)	1.33 (0.13)[a]	1.52 (0.35)[a]	1.45 (0.23)[a]	1.62 (0.34)[a]	1.67 (0.15)[a]
Detail reproduction (μm)	50	50	50	50	50

N=3 for each test group, except for Detail reproduction (N=9).
Within a row, groups superscripted with similar lower case letters showed no statistically significant differences ($P>0.05$).

Discussion

Impression materials used for making the preliminary impression for complete dentures must have a high viscosity for the anatomical forms of alveolar ridge to be properly recorded. In this study, we investigated the effect of addition of HWSA to alginate impression materials with the aim of developing a new high viscosity impression material.

The results showed that the addition of HWSA significantly increased the viscosity of the impression materials. When the alginate impression material powder is mixed with water, the calcium ions from the gypsum react with the sodium phosphate to form insoluble calcium phosphate. Calcium phosphate rather than calcium alginate is formed, because it has lower solubility; thus the sodium phosphate is called a retarder. After depletion of the retarder, the calcium ions react with the soluble alginate to form the insoluble calcium alginate, which together with water, forms an irreversible gel. The viscosity of the impression materials before the start of the gelation simply depends on the viscosity of the soluble alginate itself. In short, the viscosity of the alginate sol increases with increasing molecular weight of the alginate. Thus, the addition of HWSA causes an increase in the viscosity of the material. In addition, a significant and positive relationship was observed between the percentage of addition of HWSA and the viscosity. Moreover, the coefficient r[2] was high in the present study, indicating the predictability of the viscosity from the percentage of addition of HWSA. In other words, the viscosity of the alginate impression material can be increased or decreased by applying regression models.

The addition of HWSA may adversely affect the setting time and physical properties. The gelation process involves reaction of the sodium alginate with the sparingly soluble calcium sulfate to form the insoluble calcium alginate gel, a fibrillar cross-linked substance. In this reaction, the calcium ions replace the sodium ions in two adjacent sodium alginate molecules, cross-linking the two molecules. Calcium sulfate supplies calcium ions at a slower rate so that only a fraction of the alginate molecules become cross-linked, and the remaining unreacted sodium alginate sol, excess water, filler particles, and reaction by-products are enclosed in the cross-linked network.[8] Therefore, more unreacted sol and excess water would be enclosed when molecules of high molecular weight become cross-linked. Thus, the addition of HWSA may adversely affect the setting time and physical properties of the material after setting; further investigations must be conducted to address this issue.

The setting times of all the materials tested in this study were found to be within the specification, and the differences between ALF and the experimental materials were not significant. Alginate impression materials enter the gel state when approximately 10% of the carboxylate groups on the alginate chain are bound to calcium ions.[14] Since the materials showed similar setting times, the addition of HWSA had little effect on the reaction rate between the sodium alginate and the calcium ions. In short, the addition of HWSA has little effect on the setting time.

Recovery from deformation indicates the ability of the impression material to recover after it has been deformed during removal from the mouth. The greater the recovery from deformation, the more accurate the impression material is.[15] In this study, recovery from deformation of all the tested materials was found to be

within the specified limit. EM2; containing 75% of HWSA showed a significantly higher value of recovery from deformation than ALF. On the other hand, no significant differences were found among ALF, EM1, EM3 and EM4. These findings indicate that alginate molecules produce cross-linking more effectively, and form a densely arranged network when the material contains 75% HWSA and 25% sodium alginate. Furthermore, since none of the experimental materials had significantly lower values than ALF, the addition of HWSA does not appear to reduce the recovery from deformation of the alginate impression material.

Strain in compression of all the tested materials was within the specified limit. The property of strain in compression is related to the flexibility and stiffness of the material. Impression materials with high strain in compression can be removed from the mouth without injury to the tissues and can resist breakage when the set model material is removed from the impression. Meanwhile, impression materials with excessively high values of strain in compression could be expected to be deformed by the weight of the dental stones during cast construction. Therefore, it has been reported that the optimum range of strain in compression is 15.0-17.0%.[16] In the present study, the strain in compression of all the tested materials was within this optimum range, except for that of EM4. The strain in compression of EM4 was 18.0%, slightly higher than the aforementioned range. However, no significant differences were found among the tested materials in terms of the strain in compression. These results indicate that the addition of HWSA has little influence on the strain in compression of the alginate impression material.

The compressive strength of all the tested materials was also found to be within the specified limit. Sufficient compressive strength is needed because the material must resist compressive failure in any area where there is an undercut close to the impression tray and the material is compressed against the side of the tray and the tooth.[17] In this study, EM1 and EM2 had significantly higher values of compressive strength than ALF, with EM2 showing the highest value. On the other hand, no significant differences were found among ALF, EM3 and EM4. These results also indicate that EM2 showed more effective cross-linking than any of the other experimental materials. Furthermore, since none of the experimental materials had significantly lower values than ALF, the addition of HWSA appears not to reduce the compressive strength of the alginate impression material.

Detail reproduction and surface roughness were measured to ascertain the effect of HWSA addition on the compatibility with dental stones. All the tested materials complied with the specified minimum detail reproduction of 50 μm-lines. None of the materials could reproduce the 20 μm-line, which was in accordance with the finding in previous studies that no alginate impression materials that are powder-type can reproduce a 20 μm-line.[18] The surface roughness of all the experimental materials determined in this study were approximately equal to those reported in past studies,[19-21] and not significantly different from the surface roughness of ALF. The results of these two tests indicate that the addition of HWSA has little effect on the compatibility of the materials with dental stones.

The measurement results confirmed that the addition of HWSA did not adversely affect the setting time or physical properties of the materials, and what is more, addition of the appropriate amount could lead to improvement in some properties such as recovery from deformation and the compressive strength.

Conclusion

The addition of HWSA to the alginate impression material increased the viscosity of the material without deteriorating the properties of the material itself.

The viscosity of the impression material could be controlled by changing the percentage of addition of HWSA.

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